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## II.

# RADIOACTIVITY.

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The study of the discharge of electricity through gases and the properties of radioactive substances has done much to broaden our knowledge of the relations of electricity and matter. It has served to throw a new light on the ultimate constitution of matter itself, and, while confirming the older theory of a discontinuous or atomic structure, has led to the presumption that the chemical atom is not only divisible into still smaller entities, but that in some cases it can undergo a spontaneous disruption accompanied by the ejectment of certain of its constituent parts at high velocities. All this has opened a broad and attractive field for more or less legitimate speculation and conjecture.

Since the first recognition by Becquerel in 1897 of the radioactive phenomena exhibited by the element uranium, the extension of our knowledge of the radioactive substances has steadily and progressively advanced. This development has been due in great part to the early formulation of the theory of atomic disintegration, proposed in 1902 by Rutherford and Soddy, which has served as a systematic foundation and has afforded an orderly basis for the interpretation of the otherwise somewhat complicated relations.

According to this hypothesis the atoms of the primary radioactive elements are considered to undergo spontaneous disintegration and in this manner to give rise to a series of successive radioactive products, differing from the parent substances as well as from one another in physical and chemical properties and in the relative stability of their atomic systems. Simultaneously with the disruption of the atoms certain characteristic radiations are emitted by the systems, and it is these radiations which led to the discovery of the radio-elements and which particularly distinguishes them from all other types of matter.

The characteristic radiations emitted by radioactive substances are three in number and are known as the alpha, the beta and the gamma rays.

For our knowledge of the alpha radiation we are indebted chiefly to the work of Rutherford and his associates, which has shown conclusively that these rays consist of discreet particles of atomic dimensions, projected with high velocities and bearing a positive charge. The earlier investigations were conducted with a view to determining the mass of the particles from the deflections suffered by the rays in electric and magnetic fields of known strengths. A value for the ratio of the charge to the mass ($e/m$) was obtained in this manner and this led to the conclusion that, if the charge on a particle was the same as that carried by the hydrogen ion in electrolysis, the mass of the particles was approximately twice that of the hydrogen atom. Strong evidence was also obtained that the alpha particles emitted by the different radio-elements are identical in nature irrespective of the character of the particular radio-element from which they originate.

A very ingenious experiment was then devised by Rutherford and Geiger, in which a known fraction of the alpha particles emitted by a radioactive source was allowed to enter a small ionization chamber containing a gas at low pressure. Under the influence of a strong electric field the ions formed by the entering particles acquired so high a velocity that additional ions were produced by their collision with neutral gas molecules and the electrical effect was increased to a point where the action of a single particle could be readily detected. It was shown, moreover, that each of the scintillations appearing on a screen of Sidot's blende placed in the path of the rays corresponded to the impact of a single alpha particle.

In this manner the actual number of alpha particles emitted by a radioactive substance was accurately counted, and it was found that one gram of radium itself emitted $3.4 \times 10^{10}$ alpha particles per second.

By measuring the electric charge imparted to an insulated plate by the impact of a given number of alpha particles, the charge carried by each particle could be readily determined. This was found to be equal to $9.3 \times 10^{-10}$ electrostatic units. It was then shown in an ingenious manner that this charge was twice that carried by an electron or by a hydrogen ion, although preceding determinations of the latter magnitude had indicated that its value was somewhat less than one half of $9.3 \times 10^{-10}$. The recent determinations made by Millikam of the charge on an ion have shown, however, that $4.65 \times 10^{-10}$ is not far from correct and have confirmed the conclusion reached by Rutherford and Geiger that the charge on an alpha particle is equal to twice the unit charge of the hydrogen atom in electrolysis.

With this modification, the ratio of the charge to the mass of an alpha particle indicates that the mass is equivalent to an atomic weight of four. This corresponds to the mass of an atom of the gaseous element helium.

The final proof of the intimate connection of the alpha particle with the helium atom was supplied by Rutherford and Royds who proved by spectroscopic methods that readily detectible amounts of helium could always be obtained when large numbers of alpha particles were allowed to penetrate through a thin glass wall into a highly evacuated receptacle or into a screen of lead, from which the helium was ultimately released by fusion of the metal.

It is a significant fact that although the alpha particles from the different types of radioactive matter appear to be all of a similar nature and to consist of atoms of helium bearing a double, positive charge, the velocities at which they are ejected are different for the different radioactive substances. The velocity of the particles emitted by the atoms of any one type of matter undergoing transformation is, however, always the same and is a characteristic constant for that particular radio-element. Attention was first called to this important relation by Bragg and Kleeman and it is undoubtedly significant in its bearing on the constitution of the radio-atoms. The observed velocities of the particles appear to lie between the limits of $1.5 \times 10^{9}$ and $2.25 \times 10^{9}$ centimeters per second.

Owing to their high velocities the alpha particles are capable of passing through thin layers of ordinary matter, and can penetrate into air at atmospheric pressure for distances of from somewhat less than three to about eight centimeters. They produce phosphorescence and chemical action in substances on which they impinge and strongly ionize gases through which they pass.

Through measurements of their deflection in electric and magnetic fields the beta rays emitted by radioactive substances have been shown to consist of negatively electrified particles or electrons with an apparent mass about one eighteen-hundredth that of the hydrogen atom. The velocity with which they are projected is considerably higher than that of the alpha particles and in some cases exceeds nine tenths the velocity of light. They are capable of penetrating through moderate thicknesses of ordinary matter and for considerable distances in air. They cause phosphorescence and chemical action in substances on which they fall and produce ions in gases through which they pass.

Although the beta particles emitted by the different types of radioactive matter are in all respects identical in nature, they exhibit the same peculiarities with respect to the velocities with which they are initially projected that has been observed in the case of the alpha rays. The velocity of the beta rays from any given radio-element is the same within certain limits for every disintegrating atom of that element, but is different from the velocity of the rays emitted by other elements. The velocity of the beta rays is therefore characteristic for each of the substances which give rise to this type of radiation and has undoubtedly a significant bearing on the constitution of the radio-atoms. It appears probable from some recent experiments performed by Hahn and von Beyer, in which a magnetic spectrum of the beta rays emitted by certain radioactive substances was obtained, that the transformation of some of the radio-atoms is accompanied by the expulsion of a series of beta particles of different velocities. These results are very suggestive of the model atom devised by Sir J. J. Thomson in which the atom was assumed to be built up of concentric shells of electrons revolving with different velocities in circular orbits. It would seem quite possible that in such a system a rearrangement of the parts might result in the expulsion of electrons from several layers simultaneously.

The third type of radiation associated with radioactive transformations, known as the gamma rays, is similar to the X-rays and is supposed to consist of electromagnetic pulses in the ether. A rather ingenious corpuscular theory as to the nature of these rays has been proposed by Bragg, but has not met with general acceptance owing to the fact that it appears to be not altogether in accord with the experimental evidence.

The origin of gamma rays seems to be very intimately connected with the emission of beta particles, for the two types of radiation have been observed to appear simultaneously and bear a certain definite relation to one another. When the expelled beta particle has a high velocity the gamma ray is of a very penetrating character, while if the velocity of the beta particle is low the gamma ray emitted has but little power of penetrating ordinary matter.

The disintegration of the radio-elements takes place according to a very simple law and the number of atoms of any radio-element which undergo transformation in the unit time is a definite proportion of the total number of atoms initially present. The number of atoms $N$ remaining unchanged after any time $t$ is given by $N = No e^{-\lambda t}$, where $No$ is the initial number of atoms and $\lambda$ is the fraction undergoing transformation in the unit of time. This fraction has a fixed and invariable value for each separate radio-element and for this reason is known as the *constant of change* for the element in question. It is a relatively simple matter to determine the period of time required under these conditions for exactly half of any given quantity of a radio-element to be converted into other substances and this time is known as *the half value period.* The rate of change and the corresponding half value period is a definite characteristic for each of the radio-elements but is very different for the different radioactive substances. The half-value period of uranium, for example, is over five billion years while the half-value period of certain other radio-elements is only a few seconds.

Although the disintegration of some of the radio-elements has been examined over wide extremes of temperature and pressure, and under various other special conditions which would greatly influence the course of ordinary chemical reactions, it has not been found possible to definitely alter or effect the rate at which transformation takes place to the slightest measurable degree. It is therefore evident that the disintegration of the radio-active substances is of a wholly different character from the ordinary chemical changes. This is exactly what would be expected if the radioactive processes occur within the atoms themselves, for, in accordance with our general theories, chemical forces appear to be restricted in their action to the exterior of the atomic systems only.

We have thus far considered only the laws which govern the transformation of radioactive matter and the radiations which accompany the disintegration of the atoms; let us now turn our attention to the substances themselves. Investigation has brought to light three main groups of radioactive elements—the uranium series, the thorium series and the alkali metals. Of the last mentioned our knowledge is not very extensive. A type of beta radiation appears to be emitted by the salts of potassium and rubidium but their title to be considered as true radio-elements is not as yet entirely clear.

The uranium series, in addition to the parent substance, contains ten products which may be properly considered as in the main line of descent. These are uranium X, ionium, radium, radium emanation, radium A, radium B, radium C, radium D, radium E and radium F. Each of these products exhibits a characteristic chemical behavior which is different from that of the parent element uranium. The half value period of uranium has already been mentioned as exceeding five billions of years and the disintegration of its atoms is accompanied by the expulsion of alpha particles. Uranium X, ionium and radium are solids, the two first having chemical properties similar to thorium, while radium has those of an alkali earth and particularly resembles barium. Uranium X has a half value period of about twenty-four days, and it emits only beta and gamma rays. The rate of disintegration of ionium is not as yet known with any degree of accuracy but it is certainly a relatively stable product and is transformed but slowly. Its half value period is probably of the order of ten thousand years. It emits alpha rays only. The half value period of radium is approximately two thousand years. Rutherford and Geiger have deduced a somewhat lower value, namely 1,760 years, as a result of their experiments, but this value is probably an under estimate, as will be explained later in this paper. Radium emits alpha rays and probably very low velocity beta rays also. The change following radium is a striking one for the product in this case is gaseous. It is known as the radium emanation and has the inert chemical character of the rarer gases of the atmosphere, helium, neon, argon, krypton and xenon. When the atoms of radium emanation undergo transformation, the succeeding product known as radium A is formed. This is a solid and is deposited in the form of a thin coating of active matter on the walls of a vessel containing the emanation. This acquirement of activity by the surface of objects in contact with the emanation was observed some time before a satisfactory explanation of the phenomenon was suggested. It was therefore known as "imparted" or "induced" activity. It is now called the active deposit. Radium A, which has a half value period of three minutes and emits alpha rays, is followed by radium B, which emits beta rays and is half transformed in about twenty-six minutes, and this in turn is succeeded by radium C with a half value period of about nineteen minutes. The transformation of radium C is accompanied by the expulsion of alpha, beta and gamma rays. An interesting product known as radium D then ensues, its transformation being characterized by the absence of any detectible radiation whatever. A product of this sort is known as a *rayless change* and other examples to such a change occur in both the thorium and actinium series. On account of the similarity of its chemical properties to those of ordinary lead, radium D is known as radio-lead. It undergoes transformation more slowly than the immediately preceding products and has a half value period of about sixteen years. It is followed by radium E, a beta ray change, with a period of five days, and this is succeeded by radium F, otherwise known as polonium. Polonium emits alpha rays only and is half transformed in one hundred and forty-three days.

In addition to the ionium-radium series, uranium is also the progenitor of another group of radio-elements of which actinium is the first and most stable member. The rate of change of actinium has not yet been determined, but is comparatively slow and is probably of the same order as that of the radium. Actinium offers another example of the rayless changes which have already been referred to, and no radiations have been observed to accompany its transformation.

Six products subsequent to actinium have thus far been identified in this series. The first is radioactinium, an element having a half-value period of 19½ days and emitting both alpha and beta particles. The subsequent product is known as actinium X. The half value period of actinium X is about ten days and its atoms disintegrate with the expulsion of alpha particles. The next step in the series of transformation is the gaseous product known as the actinium emanation. This, like the radium emanation, is chemically inert and incapable of entering into combination with other elements. Actinium emanation is a very short lived substance and has a half value period of only 3.9 seconds. It is transformed successively into three other products, which are solids, known respectively as actinium A, B and C, and together constitute the so-called active deposit from the actinium emanation. Actinium A has a half value period of 36 minutes, actinium B of 3.1 minutes and actinium C of 5.1 minutes. The first emits beta rays, the second alpha rays, and the third beta and gamma rays.

As already stated, actinium and its products are genetically connected with uranium and are, in some manner as yet obscure, derived from it. The evidence in support of this conclusion is quite convincing. All uranium minerals contain definite quantities of actinium and in the older minerals the relative proportions of uranium and actinium present are so constant as to permit of no other explanation. But the actual genealogical history of actinium is still obscure and we are not yet in a position to clearly trace the line of descent. Whether actinium is formed directly from uranium by a special kind of transformation which involves only a small proportion of the total number of the atoms changing, or whether its production occurs at a later stage in the uranium-radium series, is at present an open question and the discovery of the true relations is one of the most interesting problems now awaiting solution.

The thorium series presents another group of radio-elements comprising ten successive members. I shall not stop to enumerate these in detail, but their principal physical and chemical characteristics have already been determined. Thorium itself, the parent substance, has a very slow rate of change, which is probably not more than a fifth that of uranium.

Its first product, mesothorium 1, is another example of a rayless change like radium D and actinium. Owing to the fact that large quantities of monazite are commercially treated for the extraction of thorium used in the manufacture of incandescent gas mantles and that the technical separation and isolation of mesothorium appears to be an economic possibility, there is some prospect that mesothorium may become a competitor of radium for scientific and therapeutic uses. Its life compared with radium is relatively short, however, its half value period being 5½ years, but this in itself is not necessarily a serious disadvantage. The chemical properties of mesothorium are similar to those of radium and barium.

The fifth product in the thorium series is known as the thorium emanation and is a chemically inert gas like the radium and actinium emanations. The remaining four products constitute the *thorium active deposit.*

The combined uranium and thorium series includes 28 radio-elements, of which only the two parent elements were known before the development of radioactive methods. Radioactivity has therefore added a considerable quota to the known types of matter.

An interesting relation which is met in the study of radioactive change is the so-called *radioactive equilibrium.* If a relatively long-lived radio-element A is the parent of a less stable product B, and if A is initially entirely freed from B, then a certain definite fraction of the atoms of A will undergo transformation each second to form atoms of the product B. The number of atoms of B produced from A in this manner each second will be essentially constant and the amount of B will increase. But the atoms of B also undergo transformation at a constant rate and, as the quantity of B increases, a continually increasing number of its atoms will be transformed in the unit of time. A point will finally be reached where the number of atoms of B which disintegrate in any given time will be exactly equal to the number of atoms of B formed from A in the same interval. The relative amounts of A and B will then remain constant and the conditions can be expressed by the equation

$$\lambda_1 P = \lambda_2 Q,$$

where $P$ is the number of atoms of $A$ and $\lambda_1$ its constant of change, and $Q$ is the number of atoms of $B$ and $\lambda_2$ the constant of change of the product B. Under these conditions the substances A and B are said to be in *equilibrium* with one another. The general mathematical theory of successive changes of this kind has been developed by Rutherford.

In the discussion of the characteristics of the alpha rays it has been pointed out that the evidence supplied by the determination of the ratio of the charge to the mass of these particles indicates that their nature is the same in all cases. Let us consider briefly the additional facts which are in support of this conclusion.

The presence of considerable proportions of helium in crystalline minerals containing uranium and thorium has been very frequently noticed. It was found by Ramsay and Soddy in 1903 that helium could be detected in the residual gas set free when a specimen of crystalline radium bromide was dissolved in water, and shortly after this they showed that the spectrum of helium appeared with time in a tube which initially contained only radium emanation. Debierne found that helium was produced by a strong preparation of actinium, and conclusive proof has also been obtained by Strutt and by Soddy that helium results from the disintegration of both thorium and uranium.

During the past year I was able to experimentally demonstrate the production of helium by ionium, and some earlier experiments carried out by Professor Rutherford and myself showed that helium appeared during the disintegration of polonium also. The latter conclusion has since been confirmed by the work of Mme. Curie and Debierne.

The data supplied by the counting experiments of Rutherford and Geiger afford a basis for the calculation of a number of important physical quantities, such as the mass of the hydrogen atom, the number of atoms in one gram of hydrogen and the number of molecules per cubic centimeter of any gas at standard pressure and temperature. In a similar matter Rutherford and Geiger have calculated the amount of helium produced per year by one gram of radium containing equilibrium amounts of its three alpha ray products, the emanation, radium A and radium C. The number obtained in this way was 158 cubic millimeters of helium per year per gram of radium. Measurements of the rate of production of helium by a radium salt have been carried out by Sir James Dewar and have given results somewhat in excess of this, namely 182 and 169 cubic millimeters. As a confirmation of the accuracy of Rutherford and Geiger's values, however, it may be stated that an investigation of the production of helium by radium made last year by Professor Rutherford and myself gave results in excellent agreement with the calculated value. An account of these experiments will be published shortly.

In connection with these results there is, however, one rather important point which should be mentioned. This is the fact that the rate of production of helium and the half value period of radium as calculated by Rutherford and Geiger from the results of their counting experiments, are directly dependent of the purity of the salt of radium used as a standard in their measurements. They assumed that the material of their radium standard was pure anhydrous radium bromide containing 58.5 per cent. of radium. If this was not the case and the material used as their standard contains less than the theoretical amount of radium, their calculation of the number of alpha particles emitted per gram of radium and the rate of production of helium is too low, and their estimate of the half value period of radium is too high. If, on the other hand, the material of their standard consists in part of some other compound of radium containing a higher proportion of this element than is contained in the bromide, their value for the number of alpha particles emitted and the rate of production of helium is too high and their calculated rate of disintegration of radium is too low.

There are certain reasons which lead me to believe that the radium standard used by Rutherford and Geiger actually contains a higher proportion of the element radium than they have assumed in their calculation, and that the true half value period of radium is greater than 1,760 years as they have deduced it. In 1908 I published an account of some experiments on the growth of radium in ionium preparations, which pointed to two thousand years as the half-value period of the former. This estimate was quite independent of any radium standard and I am of the opinion that it is nearer the true value than is the estimate made by Rutherford and Geiger.

The point can be definitely settled, however, by a comparison of Rutherford's standard with a standard of indisputable purity. Such a standard is in prospect in the not distant future and its preparation has been undertaken by Mme. Curie on behalf of the International Radium Standards Committee appointed at the recent Radiological Congress in Brussels.

A very interesting action which has been observed to accompany radioactive transformations is known as the *recoil phenomenon.* When a plate bearing a thin layer of very active material is placed in close proximity of another plate which is inactive, a portion of the active matter becomes detached from the film and is deposited on the surface of the second plate.

The effect is increased considerably if the receiving plate is charged negatively with respect to the plate bearing the active coating. This action is apparently due to the fact that, when the alpha or beta rays are expelled at a high velocity from a radio-atom undergoing transformation, the reaction on the residual atom causes this to move in the opposite direction with sufficient force to detach it from the plate. The action is analogous to the recoil of a rifle attending the expulsion of a high velocity bullet. When, for example, the active coating on the first plate consists of radium A then the active matter received on the second plate is composed almost exclusively of radium B; and when the film consists of radium B the material thrown off is for the most part radium C. This and other similar effects which have been noted are all of such a nature as to suggest that the explanation proposed for this interesting phenomenon is the correct one. The effect of the electric field indicates that in some way these "rest atoms" acquire positive electric charges.

From the standpoint of the disintegration theory, it is evident, when we consider the three principal groups of radioactive substances, the uranium-radium group, the actinium group, and the thorium group, that the radioactive phenomena exhibited by the atoms abruptly disappear after they have passed through a certain series of transformations, which terminates with radium F in one instance, with actinium C in another and with thorium D in the third. The apparent explanation of this circumstance would seem to be, that, following the last active change, the residual atomic nucleus finally attains a permanently stable form which undergoes no further alterations. If such is indeed the case, then we might expect that these ultimate end products of radioactive decay would accumulate in old radioactive minerals where the process of transformation has been proceeding for long geological periods. This line of reasoning has enabled us to identify at least one of these products and that is, in all probability, the one following radium F. The residual atom in this case appears to be no other than the atom of ordinary lead. There are, moreover, certain theoretical arguments which point to the same conclusion. The accepted atomic weight of uranium is 238.5. It has been found that two alpha particles are emitted during its transformation and one by the succeeding product, ionium. This would correspond to the loss of three alpha particles or helium atoms with an atomic weight of four or a total of twelve units. Two hundred and thirty-eight and one half less twelve give two hundred and twenty-six and one half for the atomic weight of radium, which corresponds to the value obtained in the actual determination of the atomic weight of this element by Mme. Curie. The transformations of the atoms of radium, the emanation, radium A, radium C and radium F are each accompanied by the expulsion of another alpha particle, making five in all. Five times four is twenty and two hundred and twenty-six and one half less twenty is two hundred and six and one half. The latter number is sufficiently near to two hundred and seven and one tenth, the most recently determined atomic weight of lead, to support the conclusion that lead is the ultimate disintegration product of radium. It has not yet been possible to determine the end products of the actinium or the thorium series but they will undoubtedly be identified among the various elements occurring in small proportions in the older uranium and thorium minerals.

Before completing this necessarily brief résumé of the present status of the study of radioactive phenomena it is necessary to make some reference to the series of papers published by Sir William Ramsay associated with A. T. Cameron and F. L. Usher. These papers, which deal with the action of radium emanation on various other substances, suggest the occurrence of certain changes, which if they really took place would be of fundamental importance to the theory of the constitution of matter. Unfortunately, so little weight can be attached to these results and the conclusions reached by these authors, that they have received no serious consideration from those most competent to judge their value.

In closing, a point which seems worthy of special emphasis may be briefly mentioned. This is the apparently important part played by the alpha or *material* particle emitted during radioactive transformations. In those cases where it has been possible to observe its influence, the loss of an alpha particle is always accompanied by a corresponding decrease in the mass of the atom from which it is separated. Although the disruption of a radio-atom is accompanied by the release of a relatively enormous amount of energy, still it appears that the fragments projected into space are always of one or the other of two quite distinct classes; either beta particles of extremely small mass, or helium atoms of a mass seven thousand times that of the beta particles. So far as the results of our experiments have enlightened us we have not yet been able to observe the resolving of the by far the greater proportion of the *effective* mass of an atom into anything other than a further subdivision of ordinary matter.

YALE UNIVERSITY,
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